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## TRANSLATION FROM JAPANESE

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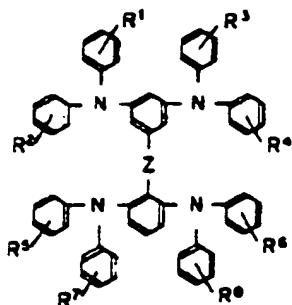
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- (21) Application No.: 3-40269  
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- 
- (54) [ Title of the Invention ] **A Phenylenediamine-Based Compound and an  
Electrophotographic Photoreceptor Manufactured Using Said Compound**  
(57) [ Summary ]  
[ Structure ]

A phenylenediamine-based compound which is represented by General  
Formula 1 below:

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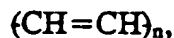
[ Chemical Formula 1 ]



(1)

(in the formula,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$ , which may be either identical or different, are each a hydrogen atom, a halogen atom, an alkyl group, or an alkoxy group; and Z is a thio group, an oxy group, a seleno group, a telluro group, an alkylene group, a group represented by the formula

[ Chemical Formula 2 ]



(in the formula,  $n$  is an integer from one to two), or a group represented by the formula

[ Chemical Formula 3 ]



(in the formula, Y is a sulfur atom, an oxygen atom, or a nitrogen atom); and an electrophotographic photoreceptor furnished with a photoreceptive layer containing said compound.

[ Merits ]

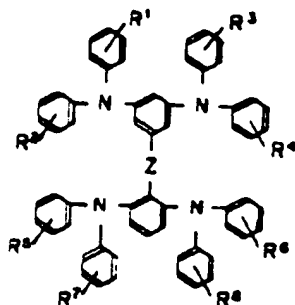
Because said compound exhibits excellent light stability, it is suitable for use as a charge-transfer material and makes it possible to obtain electrophotographic photoreceptors with excellent light stability, high sensitivity, and superb repetition characteristics.

[ Claims ]

[ Claim 1 ]

A phenylenediamine-based compound which is represented by General Formula 1 below:

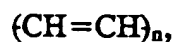
[ Chemical Formula 1 ]



(1)

(in the formula, R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup>, R<sup>5</sup>, R<sup>6</sup>, R<sup>7</sup>, and R<sup>8</sup>, which may be either identical or different, are each a hydrogen atom, a halogen atom, an alkyl group, or an alkoxy group; and Z is a thio group, an oxy group, a seleno group, a telluro group, an alkylene group, a group represented by the formula

[ Chemical Formula 2 ]



(in the formula, *n* is an integer from one to two), or a group represented by the formula

[ Chemical Formula 3 ]

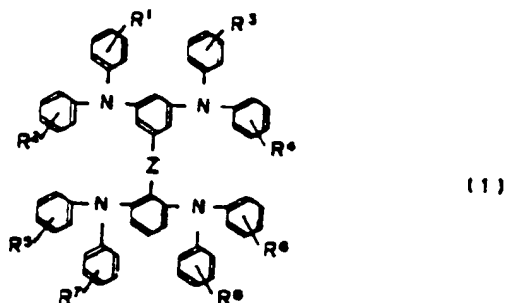


(in the formula, Y is a sulfur atom, an oxygen atom, or a nitrogen atom).

[ Claim 2 ]

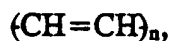
An electrophotographic photoreceptor which is characterized by the fact that a photoreceptive layer, which contains the phenylenediamine-based compound represented by General Formula 1 below, is furnished on the electroconductive substrate.

[ Chemical Formula 4 ]



(In the formula,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$ , which may be either identical or different, are each a hydrogen atom, a halogen atom, an alkyl group, or an alkoxy group; and Z is a thio group, an oxy group, a seleno group, a telluro group, an alkylene group, a group represented by the formula

[ Chemical Formula 5 ]



(in the formula,  $n$  is an integer from one to two), or a group represented by the formula

[ Chemical Formula 6 ]



(in the formula, Y is a sulfur atom, an oxygen atom, or a nitrogen atom).

[ Detailed Description of the Invention ]

[ 0001 ]

[ Field of Industrial Utilization ]

The present invention concerns a phenylenediamine-based compound suitable for use as a charge-transfer material in an electrophotographic photoreceptor, and concerns an electrophotographic photoreceptor in which said compound is used.

[ 0002 ]

[ Prior Art ]

In recent years, organic electrophotographic photoreceptors, which are easy to handle, economically efficient, and allow considerable freedom in functional design, have been widely used as electrophotographic photoreceptors in copiers and other imaging apparatuses. The Carlson process has been widely used in the formation of copy images by means of electrophotographic photoreceptors. The Carlson process involves the following steps: a discharge step during which an electrophotographic photoreceptor is charged uniformly by a corona discharge; an exposure step during which the charged electrophotographic photoreceptor is exposed to the original to form an electrostatic latent image which corresponds to the original; a development step during which the electrostatic latent image is developed with a developer containing a toner, and a toner image is formed; a transfer step during which the toner image is transferred to paper or some other medium; a fixing step during which the tone image thus transferred is fixed; and a cleaning step during which measures are taken to remove the toner that has remained on the electrophotographic photoreceptor following the transfer step. For high-quality images to be formed during the Carlson process, it is required that the electrophotographic photoreceptor have excellent charging characteristics and excellent photoreception characteristics, and that it generate only a low residual potential following exposure.

[ 0003 ]

In the past, selenium, cadmium sulfide, and other inorganic photoconductors have been known as electrophotographic photoreceptor materials, but these materials are disadvantageous because they are toxic and expensive to produce. Therefore, it has been proposed to replace these inorganic substances with so-called organic electrophotographic photoreceptors manufactured using various organic substances. An organic electrophotographic photoreceptor is furnished with a photoreceptive layer consisting of a charge-generating material, which generates a charge upon exposure, and a charge-transfer material, whose function is to transport the charge thus generated.

[ 0004 ]

The charge-generating materials and charge-transfer materials must be adequately selected if the various conditions required of such organic electrophotographic photoreceptors are to be met. Polyvinyl carbazole, oxadiazole-based compounds, pyrazoline-based compounds, hydrazone-based compounds, and various other organic compounds have been proposed and commercialized as such charge-transfer materials. Known hydrazone-based materials are described, for example, in Japanese Laid-Open Patent Applications 54-59143 and 2-210451.

[ 0005 ]

[ Problems Which the Invention Is Intended to Solve ]

However, the above-mentioned charge-transfer materials have comparatively low drift mobilities, which are a measure of the charge-transfer capability. Another drawback is the strong dependence of the drift mobility on the electric field strength, with the result that the mobility of charges in weak electric fields is low, and the residual potential is difficult to remove. Yet another drawback is that the materials are prone to degradation when irradiated with ultraviolet light.

[ 0006 ]

To overcome these drawbacks, *N,N,N',N'*-tetraphenyl-1,3-phenylenediamine has been proposed (Japanese Laid-Open Patent Application 1-142642) as an *m*-phenylenediamine-based compound with a reduced dependence of the drift mobility on the electric field strength and with satisfactory miscibility with resins. Said *m*-phenylenediamine-based compounds have satisfactory lightfastness with respect to ultraviolet light and the like, and exhibit stable characteristics when used in actual copiers. The disadvantage of these compounds, however, is that they are damaged irreversibly when exposed to light for a long time or at high temperatures during copier breakdowns or the like. Yet another disadvantage [of these compounds] is that they have insufficient sensitivity and inferior repetition characteristics.

[ 0007 ]

The objective of the present invention is to offer a phenylenediamine-based compound which exhibits excellent lightfastness and light stability and is suitable as a charge-transfer material, and to offer an electrophotographic photoreceptor in which

said compound is used and which exhibits high sensitivity and excellent repetition characteristics.

[ 0008 ]

[ Means Used to Solve the Above-Mentioned Problems, and Effect of the Invention ]

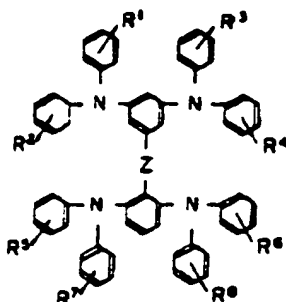
In general, the reason that photoreceptor characteristics decline as a result of photodegradation is that impurities, which serve as traps for a charge-transfer material, form in the photoreceptor. In the case of an *m*-phenylenediamine-based compound, such photodegradation reactions are believed to be the ring-closure reactions between the central benzene ring and other phenyl groups. It is believed that these reactions are facilitated by the fact that the electron density in the molecules of the phenylenediamine-based compound is shifted toward the central benzene ring. In particular, because of its configuration, position 5 on the central benzene ring has such a molecular structure that it is easily attacked by oxygen and other oxidizing substances upon optical excitation, and ring-closure reactions are believed to occur as a result of electrons being removed from this region. Therefore, it was assumed that the reactivity of a phenylenediamine-based compound can be suppressed if said region is protected by replacement with a substituent group, thus improving the stability of the compound with respect to light. As a result of various experiments, it was discovered that when a predetermined substituent group is placed in said position, the light stability of an electrophotographic photoreceptor can be effectively improved without impairing the drift mobility and other charge-transfer properties.

[ 0009 ]

Thus the phenylenediamine-based compound pertaining to the present invention is characterized by the fact that it is represented by General Formula 1 below:

[ 0010 ]

[ Chemical Formula 7 ]

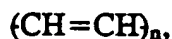


[ 0011 ]

(in the formula,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$ , which may be either identical or different, are each a hydrogen atom, a halogen atom, an alkyl group, or an alkoxy group; and Z is a thio group, an oxy group, a seleno group, a telluro group, an alkylene group, a group represented by the formula

[ 0012 ]

[ Chemical Formula 8 ]



[ 0013 ]

(in the formula,  $n$  is an integer from one to two), or a group represented by the formula

[ 0014 ]

[ Chemical Formula 9 ]



[ 0015 ]

(in the formula, Y is a sulfur atom, an oxygen atom, or a nitrogen atom). In addition, the electrophotographic photoreceptor pertaining to the present invention which is developed with the aim of attaining the stated objective is characterized by the fact that a photoreceptive layer, which contains the phenylenediamine-based compound represented by General Formula 1 above, is furnished on the electroconductive substrate.

[ 0016 ]

With the phenylenediamine-based compound represented by General Formula 1 above, position 5 on the central benzene ring is protected with a substituent group, so an attack from an oxidizing substance or the like is less likely, the ring-closure reactions are impeded, and stability with respect to light is improved. In addition, the electrophotographic photoreceptor containing the phenylenediamine-based compound represented by General Formula 1 above suffers less damage than conventional electrophotographic photoreceptors when exposed to light for a long time or at high temperatures, and thus exhibits excellent light stability.

[ 0017 ]



Another advantage is that the phenylenediamine-based compound expressed by General Formula 1 above has an excellent charge-transfer capability, so the incorporation of this phenylenediamine-based compound as a charge-transfer material into a photoreceptive layer results in a highly sensitive, easily chargeable electrophotographic photoreceptor with excellent repetition characteristics. Examples of the aforementioned halogen atoms include fluorine, chlorine, bromine, and iodine.

[ 0018 ]

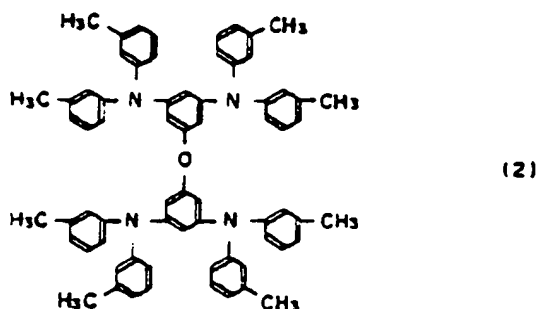
Examples of alkyl groups include such lower alkyl groups with carbon numbers from one to six as the methyl group, ethyl group, propyl group, isopropyl group, butyl group, isobutyl group, *t*-butyl group, pentyl group, and hexyl group. Examples of alkoxy groups include such lower alkoxy groups with carbon numbers in the alkyl portion from one to six as the methoxy group, ethoxy group, isopropoxy group, butoxy group, *t*-butoxy group, pentyloxy group, and hexyloxy group.

[ 0019 ]

Examples of alkylene groups include such lower alkylene groups with carbon numbers from one to six as the ethylene group, propylene group, butylene group, amylene group, and hexylene group. Examples of the phenylenediamine-based compounds represented by General Formula 1 above include the compounds represented by Formulas 2 through 9 below.

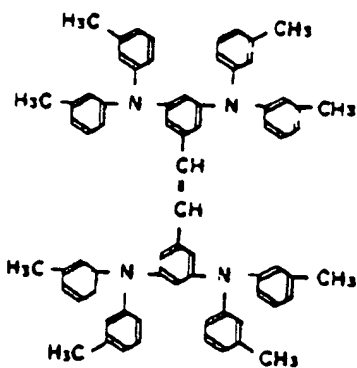
[ 0020 ]

[ Chemical Formula 10 ]

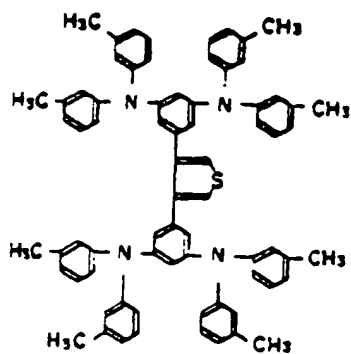


[ 0021 ]

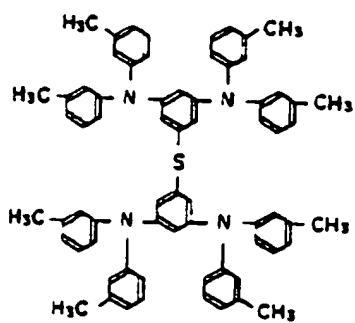
[ Chemical Formula 11 ]



(3)



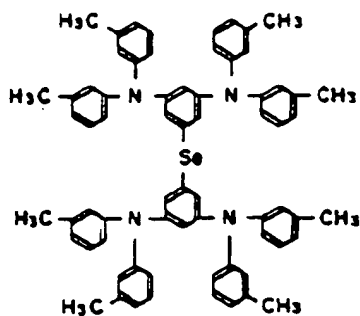
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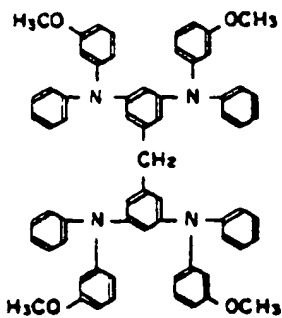
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[ 0022 ]

[ Chemical Formula 12 ]



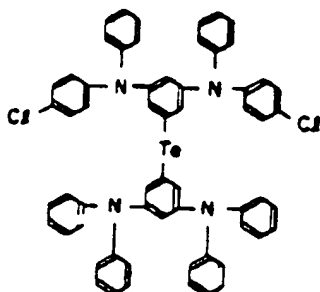
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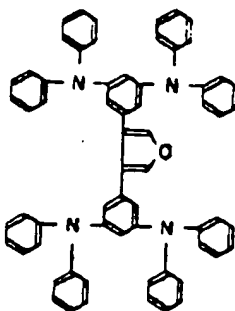
( 7 )

[ 0023 ]

[ Chemical Formula 13 ]



( 8 )



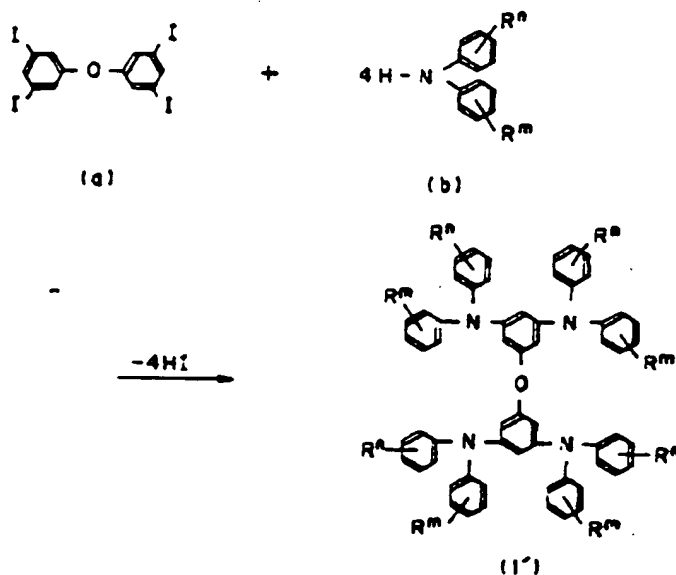
( 9 )

[ 0024 ]

The phenylenediamine-based compound pertaining to the present invention can be synthesized by various methods; for example, by employing the reaction whose formula is shown below.

[ 0025 ]

[ Chemical Formula 14 ]



[ 0026 ]

(In the formula,  $R^n$  and  $R^m$ , which may be identical or different, are each a hydrogen atom, a halogen atom, an alkyl group, or an alkoxy group). According to this reaction formula, the phenylenediamine-based compound pertaining to the present invention that is represented by Formula 1' is obtained by initiating a reaction in the presence of copper or a basic substance in an organic solvent; the reagents are the phenyl ether compound containing two substituent iodo groups that is represented by Formula (a), and the secondary amine compound containing two linked phenyl groups that is represented by Formula (b).

[ 0027 ]

The molar amount of the secondary amine compound (b) that is allowed to take part in the reaction should be at least four times the molar amount of the above-mentioned phenyl ether compound (a). NaOH,  $\text{KCO}_3$ , and  $\text{NaCO}_3$  are examples of the above-mentioned basic substances, while nitrobenzene, dichlorobenzene, *n*-methyl

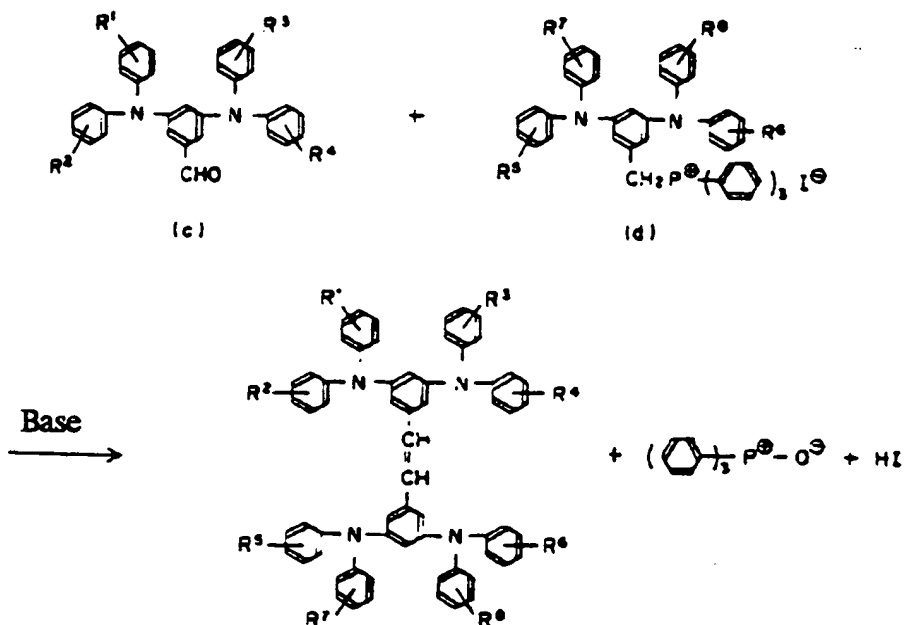
pyrrolidone are examples of the organic solvents. The reaction should be conducted at approximately 190 to 220°C.

[ 0028 ]

The above-mentioned phenylenediamine-based compound can also be obtained in accordance with the following reaction formula:

[ 0029 ]

[ Chemical Formula 15 ]



[ 0030 ]

(In the formula,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  are the same as above). In this reaction, which is based on the Wittig reaction, the phenylenediamine-based compound pertaining to the present invention that is represented by Formula 1'' is obtained by allowing the carbonyl compound represented by Formula (c) to react with the phosphonium salt represented by Formula (d) in the presence of a basic substance, for example, in the presence of  $\text{C}_6\text{H}_5\text{Li}$ ,  $\text{NaOH}$ , or some other catalyst. The reaction proceeds as a result of the above-mentioned phosphonium salt (d) releasing  $\text{HI}$  under the action of the basic substance.

[ 0031 ]

The above-mentioned carbonyl compound (c) and phosphonium salt (c) are used in equimolar amounts and can be allowed to react at approximately 10 to 50°C in an organic solvent. Nitrobenzene, THF, dioxane, or the like can be used as the organic

solvent. The photoreceptive layer pertaining to the present invention contains as the charge-transfer materials one, two, or more phenylenediamine-based compounds represented by General Formula 1 above.

[ 0032 ]

The photoreceptive layer pertaining to the present invention can be a of a single layer type or of a laminated type. The former is a mixture of a binder resin, a charge-generating material, and the compound which is represented by General Formula 1 above and which serves as a charge-transfer material, while the latter is a laminated structure consisting of a charge-generating layer and a charge-transfer layer. The photoreceptive layer pertaining to the present invention is applicable to either of these cases. To obtain a single-layer type electrophotographic photoreceptor, a photoreceptive layer containing a binder resin, a charge-generating material, the compound represented by General Formula 1 above that serves as a charge-transfer material, and other ingredients should be formed on an electroconductive substrate.

[ 0033 ]

To obtain a laminated electrophotographic photoreceptor, a charge-generating layer containing a charge-generating material should be formed on an electroconductive substrate by vapor deposition, application, or some other means, and a charge-transfer layer containing a binder resin and the compound represented by General Formula 1 above that serves as a charge-transfer material should be formed on said charge-generating layer. It is also possible to reverse the above operations and to form a similar charge-transfer layer on the electroconductive substrate and then form a charge-generating layer containing a charge-generating material by vapor deposition, application, or some other means. The charge-generating layer can also be formed by dispersing a charge-generating material and a charge-transfer material in a binder resin and applying the dispersion.

[ 0034 ]

Examples of the charge-generating materials include the conventionally used selenium, selenium-tellurium, selenium-arsenic, amorphous silicon, pyrylium salts, azo-based compounds, disazo-based compounds, phthalocyanine-based compounds, anthanthrone-based compounds, perylene-based compounds, indigo-based compounds, triphenyl methane-based compounds, threne-based compounds, toluidine-based

compounds, pyrazoline-based compounds, perylene-based compounds, quinacridone-based compounds, and pyrrolopyrrole-based compounds. To ensure that the wavelength absorption range lies within the appropriate region, these charge-generating materials may be used individually or in mixtures of two or more.

[ 0035 ]

The phenylenediamine-based compound represented by General Formula 1 above that is a charge-transfer material may be used alone or in combinations with other, conventional, charge-transfer materials. Various electron-attracting compounds or electron-donating compounds may be used as such conventional charge-transfer materials. Examples include 2,6-dimethyl-2',6'-di-*tert*-dibutyl diphenquinone and other diphenquinone derivatives; malononitrile, thiopyran-based compounds, tetracyanoethylene, 2,4,8-trinitrothioxanthone, 3,4,5,7-tetranitro-9-fluorenone, dinitrobenzene, dinitroanthracene, dinitroacridine, nitroanthraquinone, dinitroanthraquinone, succinic anhydride, maleic anhydride, and dibromomaleic anhydride.

[ 0036 ]

Examples of electron-donating compounds include nitrogen-containing cyclic compounds and condensed polycyclic compounds such as 2,5-di(4-methylaminophenyl)-1,3,4-oxadiazole and other oxadiazole-based compounds; 9-(4-diethylaminostyryl)anthracene and other styryl-based compounds; polyvinyl carbazole and other carbazole-based compounds; 1-phenyl-3-(*p*-dimethylaminophenyl)pyrazoline and other pyrazoline-based compounds; hydrazone compounds; triphenylamine-based compounds; indole-based compounds; oxazole-based compounds; isooxazole-based compounds; thiazole-based compounds; thiadiazole-based compounds; imidazole-based compounds; pyrazole-based compounds; and triazole-based compounds.

[ 0037 ]

These charge-transfer materials can be used individually or in mixtures of two or more. The binder resin is not always necessary when using polyvinyl carbazole or another such charge-transfer material capable of forming films. Various resins may be used as the binder resin in the above-mentioned photoreceptive layer, charge-generating layer, and charge-transfer layer. Examples include styrene-based polymers, styrene-

butadiene copolymers, styrene-acrylonitrile copolymers, styrene-maleic acid copolymers, acrylic copolymers, styrene-acrylic acid copolymers, polyethylenes, ethylene-vinyl acetate copolymers, chlorinated polyethylenes, polyvinyl chlorides, polypropylenes, vinyl chloride-vinyl acetate copolymers, polyesters, alkyd resins, polyamides, polyurethanes, polycarbonates, polyallylates, polysulfones, diallyl phthalate resins, ketone resins, polyvinyl butyral resins, polyether resins, and other thermoplastic resins; silicone resins, epoxy resins, phenolic resins, urea resins, melamine resins, and other cross-linking, thermosetting resins; and epoxy acrylates, urethane acrylates, and other photohardenable resins. These binder resins may be used individually or in mixtures of two or more.

[ 0038 ]

A solvent may be used to prepare a coating solution when a charge-generating layer or a charge-transfer layer is to be formed by application. Various organic solvent may be used as such solvents. Examples include methanol, ethanol, isopropanol, butanol, and other alcohols; *n*-hexane, octane, cyclohexane, and other aliphatic hydrocarbons; benzene, toluene, xylene, and other aromatic hydrocarbons; dichloromethane, dichloroethane, carbon tetrachloride, chlorobenzene, and other halogenated hydrocarbons; dimethyl ether, diethyl ether, tetrahydrofuran, ethylene glycol dimethyl ether, diethylene glycol dimethyl ether, and other ethers; acetone, methyl ethyl ketone, cyclohexanone, and other ketones; and ethyl acetate, methyl acetate, and other esters; as well as dimethyl formaldehyde, dimethyl formamide, and dimethyl sulfoxide. These solvents may be used individually or in mixtures of two or more.

[ 0039 ]

If the sensitivity of the charge-generating layer is to be improved, terphenyl, a halonaphthoquinone, acenaphthylene, or another such conventional sensitizing agent may be used together with the above-mentioned charge-generating material. Surfactants, leveling agents, and the like may also be used in order to improve the dispersibility, applicability, and other properties of the charge-transfer material or charge-generating material.



[ 0040 ]

Various electrically conductive materials can be used in the above-mentioned electroconductive substrate. Examples include aluminum, copper, tin, platinum, silver, vanadium, molybdenum, chromium, cadmium, titanium, nickel, palladium, indium, stainless steel, brass, and other individual metals; plastic materials onto which the above-mentioned metals have been vapor-deposited or laminated; and glass coated with aluminum iodide, tin oxide, indium oxide, or the like.

[ 0041 ]

The electroconductive substrate may be shaped as a sheet, drum, or the like, and may either be electroconductive itself or have an electroconductive surface. The electroconductive substrate should also exhibit a sufficient mechanical strength during use. The charge-generating material and the binder resin that comprise the charge-generating layer in a laminated electrophotographic photoreceptor can be used in a variety of ratios, but the suitable ratio is 5 to 500 parts (in this and further instances, "parts" refers to weight parts), and the ideal ratio is 10 to 250 parts, of the charge-generating material per 100 parts of the binder resin. The charge-generating layer may be of suitable thickness, but it is preferable to form it into a film with a thickness of 0.01 to 5  $\mu\text{m}$ , and 0.1 to approximately 3  $\mu\text{m}$  in particular.

[ 0042 ]

The aforementioned binder resin and the phenylenediamine-based compound (charge-transfer material) represented by General Formula 1 above that serves as the charge-transfer layer may be used in any of the numerous possible ratios as long as the transfer of charges is not impeded and the material does not crystallize, but if the charges that are generated in the charge-generating layer by illumination with light are to be transported easily, the phenylenediamine-based compound represented by General Formula 1 above should be used in a ratio of 25 to 200 parts, and preferably 50 to 150 parts, per 100 parts of the binder resin. In addition, the charge-transfer layer should have a thickness of 2 to 100  $\mu\text{m}$ , and 5 to approximately 30  $\mu\text{m}$  in particular.

[ 0043 ]

For each 100 parts of the binder resin, a single-layer type electrophotographic photoreceptor should contain 2 to 20 parts, and 3 to 15 parts in particular, of a charge-

generating material, as well as 40 to 200 parts, and 50 to 150 parts in particular, of the phenylenediamine-based compound (charge-transfer material) represented by General Formula 1 above. In addition, the thickness of the single-layer type photoreceptive layer should be 10 to 50  $\mu\text{m}$ , and 15 to approximately 30  $\mu\text{m}$  in particular.

[ 0044 ]

As long as the characteristics of the electrophotographic photoreceptor are not impaired, a barrier layer may be formed between the above-mentioned electroconductive substrate and photoreceptive layer in a single-layer type electrophotographic photoreceptor, and between the above-mentioned electroconductive substrate and charge-generating layer, or between the electroconductive substrate and the charge-transfer layer as well as between the charge-generating layer and the charge-transfer layer, in a laminated electrophotographic photoreceptor.

[ 0045 ]

When the above-mentioned charge-generating layer and charge-transfer layer are formed using an application method, a charge-generating material, a binder resin, and other ingredients should be dispersed and mixed by a conventional method using, for example, a roll mill, a ball mill, an attritor, a paint shaker, or an ultrasonic dispersion apparatus; a coating solution should be prepared; and said solution should be applied and dried using a conventional technique. As has already been stated above, the charge-generating layer may also be formed by the vapor deposition of the above-mentioned charge-generating material.

[ 0046 ]

[ Practical Examples ]

The present invention will now be described using practical examples and comparative examples.

#### Practical Example 1

(Synthesis of Phenylenediamine-Based Compound Expressed by Formula 2 Above)

2.2 g of the phenyl ether compound represented by Formula 10 below, 2.6 g of *N,N*-di(3-methyl phenyl)amine, and 0.01 g of copper powder were allowed to react for 24 hours under reflux in 50 mL of nitrobenzene at a temperature of 200°C. [The

reaction product] was separated and purified by an ordinary method, yielding the phenylenediamine-based compound (hereinafter referred to as "phenylenediamine-based compound 2") represented by Formula 2 above.

[ 0047 ]

[ Chemical Formula 16 ]



[ 0048 ]

Phenylenediamine-based compound 2 had a yield of 55% and was a white powder with a melting point of 188 to 192°C. The following results were obtained when said Compound 2 was subjected to elementary analysis:

Elementary analysis values for  $C_{66}H_{62}N_4^*$

Calculated values (%)	C 85.86	H 6.57	N 5.89
Measured values (%)	C 85.77	H 6.50	N 5.80

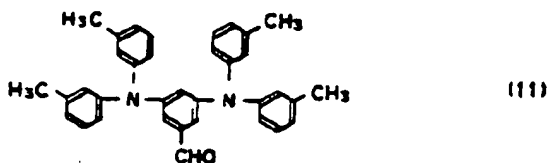
### Practical Example 2

(Synthesis of Phenylenediamine-Based Compound Represented by Formula 3 Above)

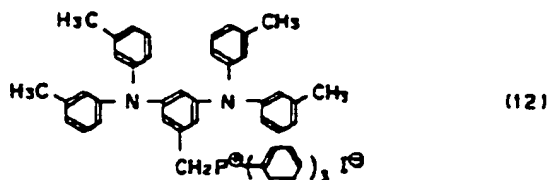
2.0 g of the carbonyl compound represented by Formula 11 below and 12.7 g of the phosphonium salt represented by Formula 12 below were allowed to react for 12 hours under reflux in 100 mL of nitrobenzene at a temperature of 20°C in the presence of 0.03 g of a basic substance ( $NaCO_3$ ), and the product was separated by recrystallization, yielding the phenylenediamine-based compound (hereinafter referred to as "phenylenediamine-based compound 3") represented by Formula 3 above.

[ 0049 ]

[ Chemical Formula 17 ]



\* Translator's note: Due to the poor legibility of the original, the small subscripts may be incorrect in this and further instances.



[ 0050 ]

Phenylenediamine-based compound 3 had a yield of 60% and was a white powder with a melting point of 179 to 182°C. The following results were obtained when said Compound 3 was subjected to elementary analysis:

Elementary analysis values for  $C_{70}H_{61}N_4$

Calculated values (%)	C 87.46	H 6.71	N 5.83
Measured values (%)	C 87.39	H 6.70	N 5.74

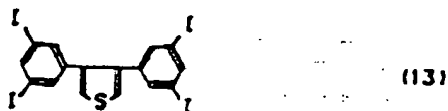
### Practical Example 3

(Synthesis of Phenylenediamine-Based Compound Represented by Formula 4 Above)

The phenylenediamine-based compound (hereinafter referred to as "phenylenediamine-based compound 4") represented by Formula 4 above was obtained in a manner similar to Practical Example 1 above, but 3.6 g of the compound represented by Formula 13 below was used instead of the phenyl ether compound represented by Formula 10 above.

[ 0051 ]

[ Chemical Formula 18 ]



[ 0052 ]

Phenylenediamine-based compound 4 had a yield of 51% and was a lemon-yellow powder with a melting point of 172 to 174°C. The following results were obtained when said Compound 4 was subjected to elementary analysis:

Elementary analysis values for  $C_{72}H_{64}N_4$

Calculated values (%)	C 85.51	H 6.38	N 5.54
Measured values (%)	C 85.44	H 6.31	N 5.53

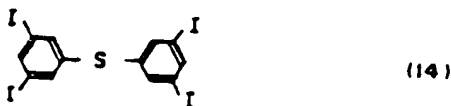
Practical Example 4

(Synthesis of Phenylenediamine-Based Compound Represented by Formula 5 Above)

The phenylenediamine-based compound (hereinafter referred to as "phenylenediamine-based compound 5") represented by Formula 5 above was obtained in a manner similar to Practical Example 1 above, but 2.48 g of the compound represented by Formula 14 below was used instead of the phenyl ether compound represented by Formula 10 above.

[ 0053 ]

[ Chemical Formula 19 ]



[ 0054 ]

This phenylenediamine-based compound 5 had a yield of 54% and was a lemon-yellow powder with a melting point of 185 to 188°C. The following results were obtained when said Compound 5 was subjected to elementary analysis:

Elementary analysis values for  $C_{66}H_{62}N_4$

Calculated values (%)	C 84.43	H 6.46	N 5.79
Measured values (%)	C 84.40	H 6.41	N 5.72

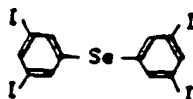
Practical Example 5

(Synthesis of Phenylenediamine-Based Compound Represented by Formula 6 Above)

The phenylenediamine-based compound (hereinafter referred to as "phenylenediamine-based compound 6") represented by Formula 6 above was obtained in a manner similar to Practical Example 1 above, but 2.6 g of the compound represented by Formula 15 below was used instead of the phenyl ether compound represented by Formula 10 above.

[ 0055 ]

[ Chemical Formula 20 ]



(15)

[ 0056 ]

Phenylenediamine-based compound 5 [sic] had a yield of 55% and was a lemon-yellow powder with a melting point of 190 to 193°C. The following results were obtained when said Compound 6 was subjected to elementary analysis:

Elementary analysis values for  $C_{66}H_{62}N_4$

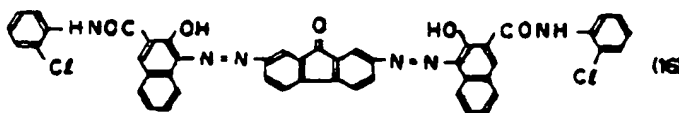
Calculated values (%)	C 80.53	H 6.16	N 5.38
Measured values (%)	C 80.49	H 6.18	N 5.36

Practical Examples 6 through 10 and Comparative Example 1 (Laminated Electrophotographic Photoreceptor)

2 parts of the compound (Compound 2) represented by Formula 16 below that served as a charge-generating material was dispersed for 2 hours together with 1 part of a polyvinyl butyral resin (BH-5, manufactured by Sekisui Chemical) and 120 parts of tetrahydrofuran in a paint shaker using zirconia beads (diameter: 2 mm). The dispersion solution thus obtained was applied to an aluminum sheet using a wire bar and dried for one hour at 100°C, yielding a 0.3  $\mu\text{m}$  charge-generating layer.

[ 0057 ]

[ Chemical Formula 21 ]



[ 0058 ]

A solution, in which 1 part of charge-transfer material and 1 part of poly(4,4'-cyclohexylidene diphenyl)polycarbonate (Z-200, manufactured by Mitsubishi Gas Chemical) had been dissolved in 9 parts of toluene, was applied with a wire bar to said charge-generating layer and dried for one hour at 100°C, yielding a 20  $\mu\text{m}$  charge-transfer layer. Table 1 shows the charge-transfer materials used in Practical Examples 6 through 10 and in Comparative Example 1. Compounds 2 through 6, which are the charge-transfer materials shown in Table 1, refer to phenylenediamine-

based compounds 2 through 6, respectively, while the comparative compound refers to *N,N',N'*-tetrakis(3-methyl phenyl)-1,3-diaminobenzene.

[ 0059 ]

Practical Examples 11 through 15 and Comparative Example 2 (Single-Layer Type Electrophotographic Photoreceptor)

1 part of the compound (Compound 1) represented by Formula 16 above that served as a charge-generating material was dispersed for 2 hours together with 60 parts of tetrahydrofuran in a paint shaker using zirconia beads (diameter: 2 mm). 10 parts of a charge-transfer material and 50 parts of a tetrahydrofuran solution of poly(4,4'-cyclohexylidene diphenyl)carbonate (Z-200, manufactured by Mitsubishi Gas Chemical) with a solid fraction content of 20 wt % were added to the dispersion solution thus obtained, and dispersion operations were continued for another hour. The dispersion solution thus obtained was applied to an alumite-treated aluminum tube and dried for one hour at 100°C, yielding a 20  $\mu$ m photoreceptive layer. The charge-transfer materials that were used in the process are shown in Table 2. The comparative compound and compounds 2 through 6 shown in Table 2 are the same as those shown in Table 1.

[ 0060 ]

(Evaluation Tests)

The following parameters of the electrophotographic photoreceptors obtained in Practical Examples 6 through 15 and in Comparative Examples 1 and 2 were measured using an evaluation testing machine (EPA-8100, manufactured by Kawaguchi Denki): the surface potential, SP (V); the half-decay exposure luminous energy,  $E_{1/2}$  (lux·sec); the residual potential,  $V_r$  (V); and the surface potential following exposure to light, SP (V). Measurement conditions were as follows.

[ 0061 ]

Luminous intensity:	50 lux
Exposure intensity:	1/15 s
Surface potential:	In the vicinity of $\pm 700$ V by adjusting influx current value
Light source:	Tungsten lamp
Charge removal:	200 lux
Measurements of residual potential:	Measurements were started 0.2 second after the start of exposure

[ 0062 ]

#### Measurements of Residual Potential Following Exposure

The value of the influx current was adjusted so as to set [the potential] in the vicinity of  $\pm 700$  V, [electrophotographic photoreceptors] were illuminated with light for 20 minutes at a luminous intensity of 1000 lux ("National Highlight FL," 15 W; manufactured by Matsushita Electric Industrial), and the surface potentials of the electrophotographic photoreceptors were then measured. Table 1 shows the test results obtained in Practical Examples 6 through 10 and in Comparative Example 1 (laminated electrophotographic photoreceptors), while Table 2 shows the test results obtained in Practical Examples 11 through 15 and in Comparative Example 2 (single-layer type electrophotographic photoreceptors).



[ 0063 ]  
[ Table 1 ]

Laminated electrophotographic photoreceptors

	Charge-transfer materials	Surface potential (V)	E <sub>1/2</sub> (lux·sec)	Residual potential (V)	Surface potential following illumination with light (V)
Practical Example 6	Compound 2	-692	1.11	-119	-648
Practical Example 7	Compound 3	-705	1.05	-110	-650
Practical Example 8	Compound 4	-695	1.08	-115	-635
Practical Example 9	Compound 5	-702	1.13	-120	-657
Practical Example 10	Compound 6	-700	1.00	-105	-630
Comparative Example 1	Comparative compound	-700	1.12	-120	-550

[ 0064 ]  
[ Table 2 ]

Laminated electrophotographic photoreceptors

	Charge-transfer materials	Surface potential (V)	E <sub>1/2</sub> (lux·sec)	Residual potential (V)	Surface potential following illumination with light (V)
Practical Example 11	Compound 2	+715	2.60	+162	+678
Practical Example 12	Compound 3	+702	2.49	+151	+650
Practical Example 13	Compound 4	+700	2.52	+155	+650
Practical Example 14	Compound 5	+720	2.62	+161	+679
Practical Example 15	Compound 6	+690	2.44	+148	+625
Comparative Example 2	Comparative compound	+713	2.60	+160	+552

[ 0065 ]

These test results reveal that all the electrophotographic photoreceptors (Practical Examples 6 through 15) that were prepared using the compounds pertaining to the present invention, which compounds are intended for use in charge-transfer materials, are almost identical to the conventional electrophotographic photoreceptors (Comparative Examples 1 and 2) in terms of surface potential, half-decay exposure luminous energy, and residual potential, and exhibit satisfactory values. However, stability with respect to light is considerably improved in comparison with that of a conventional electrophotographic photoreceptor. It was thus learned that electrophotographic photoreceptors in which the phenylenediamine-based compounds pertaining to the present invention are used as the charge-transfer materials retain satisfactory sensitivity, exhibit excellent repetition characteristics, display a markedly improved lightfastness, and have superb light stability.

[ 0066 ]

[ Merits of the Invention ]

Thus, the phenylenediamine-based compounds pertaining to this invention display excellent light stability and can be suitably used as charge-transfer materials, because phenyl groups are added to the nitrogen atoms attached to the central benzene ring, thus protecting the reaction sites, making any attack from oxidizing substances or the like more difficult, and suppressing the ring-closure reactions. The use of such phenylenediamine-based compounds as charge-transfer materials also makes it possible to obtain electrophotographic photoreceptors with excellent light stability, high sensitivity, and superb repetition characteristics.